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**Polyester Resin Composition Used For Calendering,
and a Sheet Using the Same**

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For: Eastman Chemical Company

9109 Eagle Hills Dr . Las Vegas NV 89134 . ph: 866-722-7990 (toll-free) . fax: 702-973-6009 . email: sales@tls-translations.com

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(71) Applicant: 000003160
Toyobo Co., Ltd.
2-8 Doshimahama 2-chome, Kita-ku, Osaka-shi, Osaka-fu
(72) Inventor: Kazunori Komatsu
c/o Toyobo General Research Institute
1-1 Katata 2-chome, Otsu-shi, Shiga-ken
(72) Inventor: Osamu Iritani
c/o Toyobo General Research Institute
1-1 Katata 2-chome, Otsu-shi, Shiga-ken
(72) Inventor: Mitsuo Nishida
c/o Toyobo General Research Institute
1-1 Katata 2-chome, Otsu-shi, Shiga-ken

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(54) [Title of the Invention]

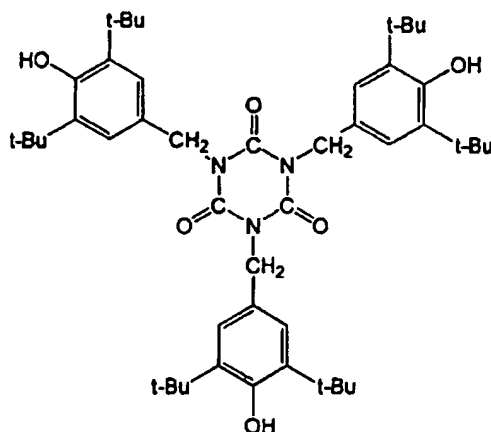
**Polyester Resin Composition Used For
Calendering, and a Sheet Using the Same**

(57) [Summary]

[Object] To provide a polyester resin composition in which the release properties of a sheet from the rolls during calendering is improved, which has extremely excellent takeoff properties in a molten sheet from the calender roll to the cooling roll, and which enables sheet calendering in which the finished sheet has good transparency and excellent quality.

[Means of Achievement] A polyester resin composition used for calendering, comprising 0.01 to 2 weight parts of the hindered phenol-type antioxidant indicated by chemical formula I and 0.01 to 5 weight parts of a lubricant per 100 weight parts of a non-crystalline polyester.

[Formula I]

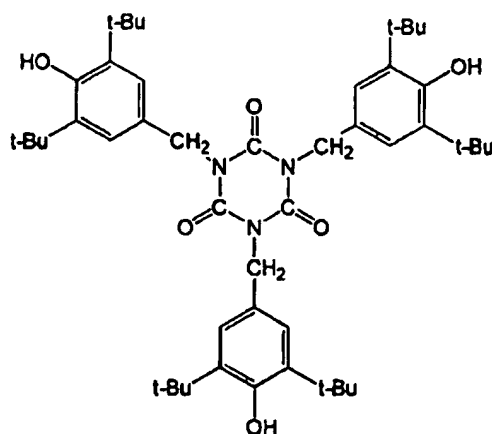


[Claims]

[Claim 1] A polyester resin composition used for calendering, characterized in comprising a non-crystalline polyester, the hindered phenol-type antioxidant indicated by chemical formula I, and a lubricant.

Formula I

[Chemical Formula 1]



[Claim 2] The polyester resin composition used for calendering according to claim 1, characterized in that the non-crystalline polyester is a copolyester that has terephthalic acid and ethylene glycol as the main components thereof, and comprises 5 to 50 mol% of isophthalic acid

with respect to the carboxylic acid component pictured previously, or comprises 5 to 50 mol% of one, two, or more types of diol components selected from the group comprising neopentyl glycol, diethylene glycol, 1,4-cyclohexanedimethanol, 2-methyl-1,3-propanediol, and 1,3-propanediol with respect to the total quantity of diol components.

[Claim 3] The polyester resin composition used for calendering according to claims 1 or 2, characterized in that the non-crystalline polyester has terephthalic acid and ethylene glycol as the main components thereof, and comprises 5 to 50 mol% of one, two, or more types of diol components selected from the group comprising diethylene glycol, neopentyl glycol, and cyclohexanedimethanol with respect to the total quantity of diol components.

[Claim 4] A sheet molded by means of calendering the polyester resin composition according to any of claims 1 through 3.

[Detailed Description of the Invention]

[0001]

[Technological Field of the Invention] The present invention relates to a polyester resin composition that is optimal for calendering; that has excellent release properties from a roll during calendering, long-run workability, thermal stability, and, particularly, takeoff properties in a calendered sheet; and that is useful in the shrink labeling used for food products, cosmetics, and beverages, as well as in aroma-retaining heat sealed film, oil-resistant multilayer sheeting, and other various sheets and films, and, particularly, in sheets and films used in industrial processes. The present invention also relates to a sheet molded from this polyester resin composition.

[0002]

[Prior Art] Vinyl chloride sheeting (film) is widely used in conventional practice in a variety of applications because of its low cost and excellent transparency. Extrusion molding and calender molding are known as methods for working this sheeting, but friction with the die lips in extrusion molding affects the workability (moldability) of the resin more than in calender molding, which lowers the precision of the sheet (film) thickness, width, flow direction, and the like, and this method is ill-suited for large-scale production. The latter method is therefore more widely employed when productivity and quality are emphasized.

[0003] Because the flexibility of vinyl chloride can be freely adjusted by means of the addition of plasticizers, it is easily workable in the form of a sheet, and an example of an application thereof is in decorative sheeting (film) imprinted with wood grain for enhancing the design of wood.

[0004] However, due to drawbacks whereby dioxins are released during incineration of vinyl chloride, and restrictions on the use of plasticizers due to the presence of endocrine-disrupting chemicals, a changeover from vinyl chloride sheeting (film) to other materials has been under way in recent years. Among substitute materials, polyester is favored for its physical aspects, cost, and other attributes. However, using polyester as a substitute has the following significant drawbacks.

[0005] One drawback is the difficulty of applying polyester to calender molding, which is widely used as a molding method for polyvinyl chloride. Extrusion molding has mainly been used conventionally as a method for manufacturing sheets and films from polyester resin for its ease of fabrication. However, molding is difficult when calendering is attempted using polyester resin, because of its relatively strong adhesion to the rolls when thermoplasticized, and its tendency to adhere to the rolls during fabrication. The addition of various lubricants has therefore been attempted in order to prevent adhesion to the rolls.

[0006] Lubricants that have thus been investigated include, for example, polyethylene wax, paraffin wax, and other hydrocarbon lubricants; higher fatty acid lubricants, higher alcohol lubricants, metallic soaps made from higher fatty acids, fatty acid amide lubricants, ester lubricants, and various other lubricants. For example, non-crystalline polyethylene terephthalate copolymerized with cyclohexanedimethanol has been made into sheets by means of calendering with the aid of various lubricants, as described in JP (Kokai) No. 11-343353, 2000-136294, 2000-186191, 2000-302951, 2001-64496, 2001-4019, and US Patent No. 6068910. The inventors and others have investigated various admixtures according to these disclosures using this polyester, but the transparency of the finished sheet declines if the quantity of lubricant is increased in order to improve roll release properties, and the proper balance between the quantity of polyester and lubricant has been impossible to achieve.

[0007] Although there are products described in these disclosures that have good release properties from a roll, significant drawbacks relating to practical application (industrial production) in these products have come to be known. Specifically, when the inventors

conducted calendering tests using the non-crystalline polyesters and lubricants disclosed therein, they discovered that although release properties from the calender roll were obtainable as the quantity of lubricant was increased, drawbacks appeared whereby takeoff from the calender rolls to the cooling rolls was unsatisfactory because of so-called "sagging" under the sheet's own weight that occurred due to insufficient strength in the molten resin itself when the sheet was taken off the calender rolls and brought to the cooling rolls.

[0008]

[Problems to Be Solved by the Invention] In view of the foregoing, an object of the present invention is to provide a polyester resin composition used for calendering that not only has good release properties of the sheet from the roll during fabrication (specifically, when making a sheet by means of a calender roll), but also good takeoff properties from the calender roll, and good transparency in the finished sheet.

[0009]

[Means Used to Solve the Above-Mentioned Problems] As a result of concentrated investigation aimed at overcoming the abovementioned drawbacks, the inventors discovered that by means of admixing a non-crystalline polyester with a specific antioxidant and lubricant, not only can good roll release properties during calendering be obtained, but a balance can also be obtained between takeoff properties in the molten sheet from the calender roll and transparency of the sheet, and thus developed the present invention.

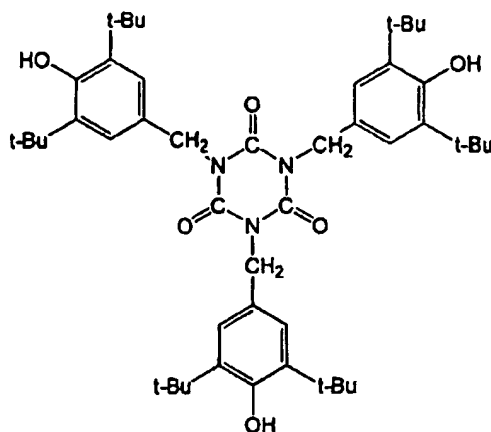
[0010] Specifically, the present invention has the following characteristics.

(1) A polyester resin composition used for calendering, characterized in comprising a polyester, the hindered phenol-type antioxidant indicated by chemical formula I, and a lubricant.

Formula I

[0011]

[Chemical Formula 2]



[0012] (2) The polyester resin composition used for calendering according to (1), characterized in that the non-crystalline polyester is a copolyester that has terephthalic acid and ethylene glycol as the main components thereof, and comprises 5 to 50 mol% of isophthalic acid with respect to the carboxylic acid component pictured previously, or comprises 5 to 50 mol% of one, two, or more types of diol components selected from the group comprising neopentyl glycol, diethylene glycol, 1,4-cyclohexanedimethanol, 2-methyl-1,3-propanediol, and 1,3-propanediol with respect to the total quantity of diol components.

[0013] (3) The polyester resin composition used for calendering according to (1) or (2), characterized in that the non-crystalline polyester has terephthalic acid and ethylene glycol as the main components thereof, and comprises 5 to 50 mol% of one, two, or more types of diol components selected from the group comprising diethylene glycol, neopentyl glycol, and cyclohexanedimethanol with respect to the total quantity of diol components.

[0014] (4) A sheet molded by means of calendering the polyester resin composition according to any of (1) through (3) above.

[0015]

[Embodiments of the Invention] The polyester resin composition of the present invention used for calendering preferably comprises 0.05 to 2 weight parts of the hindered phenol-type antioxidant indicated by chemical formula 1, and 0.01 to 5 weight parts of a lubricant per

100 weight parts of a non-crystalline polyester. "Non-crystalline" as mentioned herein is defined as follows. Specifically, using a differential scanning calorimeter (DSC), the temperature of the product is raised from -100°C to 300°C at a rate of $20^{\circ}\text{C}/\text{minute}$, the product is cooled from 300°C to -100°C at a rate of $50^{\circ}\text{C}/\text{min}$, and then the temperature is raised again from -100°C to 300°C at a rate of $20^{\circ}\text{C}/\text{min}$. The product is non-crystalline if it does not have a distinct fusion peak during either of the two temperature increases.

[0016] The non-crystalline polyester used in the present invention is not particularly limited, but a polyester in which terephthalic acid and ethylene glycol are the main components is preferred in terms of elongation and mechanical properties (particularly strength and impact resistance) of a calendered sheet thereof. A "main component" herein is considered to comprise 50 mol% or more, preferably 60 mol% or more, and more preferably 65 mol% or more of each the dicarboxylic acid component and the glycol component when the total quantity of both of these components is taken to be 100 mol%.

[0017] Furthermore, in the case of a polyester having terephthalic acid and ethylene glycol as the main components thereof, if the copolymer composition comprises (1) 5 to 50 mol% (preferably 10 to 40 mol%) of isophthalic acid components with respect to the total quantity of dicarboxylic acid components and/or (2) 5 to 50 mol% (preferably 10 to 40 mol%) of at least one type of diol component selected from the group comprising 1,2-propanediol, 1,3-propanediol, 1,4-butanediol, 2-methyl-1,3-propanediol, neopentyl glycol, diethylene glycol, and 1,4-cyclohexanedimethanol with respect to the total quantity of diol components, the crystallinity of the resin decreases, and the workability of the resin composition of the present invention is enhanced even further. Also, in the case of (2), a copolymer composition comprising one type or two or more types of diol components selected from the three diol components neopentyl glycol, diethylene glycol, and 1,4-cyclohexanedimethanol is more preferred.

[0018] Preferred effects with respect to transparency are obtained using a copolyester such as this, because the resin can be rendered non-crystalline while maintaining the good strength, elongation, and impact resistance exhibited by polyethylene terephthalate.

[0019] Also, in the case of a copolyester that satisfies (2) above, if the copolymer composition comprises neopentyl glycol, the compatibility of such a polyester with the lubricant (particularly,

the hereinafter described metal salt of an organic¹ phosphoric acid ester and polyolefinic wax) is enhanced, and the quantity of lubricant needed to obtain roll release properties can be reduced.

As a result, not only is such a composition useful in enhancing the transparency of the sheet, but mechanical characteristics such as the elongation and strength of the sheet are also enhanced.

[0020] Polycarboxylic acids other than the abovementioned terephthalic acid and isophthalic acid may be copolymerized in the non-crystalline polyester resin of the present invention, and examples thereof include orthophthalic acid, naphthalene dicarboxylic acid, succinic acid, adipic acid, azelaic acid, sebacic acid, decane acids, dimer acids, cyclohexane dicarboxylic acid, trimellitic acid, and other known products; and one, two, or more types thereof may be used.

[0021] The non-crystalline polyester resin of the present invention may be copolymerized with other polyhydric alcohol components other than the abovementioned ethylene glycol, diethylene glycol, neopentyl glycol, and cyclohexanedimethanol, and examples thereof include 1,2-butanediol, 1,3-butanediol, 1,4-butanediol, 1,5-pentanediol, 3-methyl-1,5-pentanediol, 2-methyl-1,3-propanediol, hexanediol, nonanediol, dimer diols, ethylene oxide adducts and propylene oxide adducts of bisphenol A, polyethylene glycol, polypropylene glycol, polytetramethylene glycol, 2-butyl-2-ethyl-1,3-propanediol, tricyclodecane dimethanol, hydroxyypivalic acid neopentyl ester, 2,2,4-trimethyl-1,5-pentanediol, trimethylolpropane, and the like; and one, two, or more types thereof may be used.

[0022] Specific examples of optimal non-crystalline polyesters in the present invention include terephthalic acid/isophthalic acid/ethylene glycol = (90 to 70)/(10 to 30)//100 mol%, terephthalic acid/ethylene glycol/1,2-propylene glycol = 100/(80 to 50)/(20 to 50) mol%, terephthalic acid/isophthalic acid/ethylene glycol/1,3-propylene glycol = (95 to 80)/(5 to 20)/(90 to 70)/(10 to 30) mol%, terephthalic acid/isophthalic acid/ethylene glycol/1,4-butanediol = (95 to 70)/(5 to 30)/(90 to 50)/(10 to 50) mol%, terephthalic acid/ethylene glycol/2-methyl-1,3-propanediol = 100/(60 to 80)/(4.0 to 20) mol%, terephthalic acid/isophthalic acid/ethylene glycol/2-methyl-1,3-propanediol = (95 to 80)/(5 to 20)/(70 to 90)/(30 to 10) mol%, terephthalic acid/ethylene glycol/neopentyl glycol = 100/(85 to 60)/(15 to 40) mol%, terephthalic acid/isophthalic acid/ethylene glycol/neopentyl glycol = (95 to 80)/(5 to 20)/(90 to 70)/(10 to 30) mol%, terephthalic acid/ethylene glycol/diethylene glycol = 100/(75 to 50)/(25 to

¹ Translator's note: Erroneous characters are used in the word "organic" in the original, which appears to be a typographical error. This word is corrected in the translation.

50) mol%, terephthalic acid/isophthalic acid/ethylene glycol/diethylene glycol = (95 to 80)/(5 to 20)/(90 to 75)/(10 to 25) mol%, and terephthalic acid/ethylene glycol/1,4-cyclohexane dimethanol = 100/(80 to 60)/(20 to 40) mol%. If the non-crystalline polyester consists of one of these, the transparency of a fabricated sheet with moderate crystallinity is further increased, and the glass transition temperature can be adjusted in a range such as shown below.

[0023] Furthermore, a non-crystalline polyester comprising terephthalic acid/ethylene glycol/neopentyl glycol = 100/(85 to 60)/(15 to 40) mol%, terephthalic acid/isophthalic acid/ethylene glycol/neopentyl glycol = (95 to 80)/(5 to 20)/(90 to 70)/(10 to 30) mol%, terephthalic acid/ethylene glycol/diethylene glycol = 100/(75 to 50)/(25 to 50) mol%, terephthalic acid/isophthalic acid/ethylene glycol/diethylene glycol = (95 to 80)/(5 to 20)/(90 to 75)/(10 to 25) mol%, or terephthalic acid/ethylene glycol/1,4-cyclohexanedimethanol = 100/(80 to 60)/(20 to 40) mol% can be adjusted to a high degree of balance between roll release properties, takeoff properties of the molten sheet from the calender roll, and transparency of the finished sheet, and is therefore more preferred.

[0024] The number-average molecular weight of the non-crystalline polyester used in the present invention is preferably a minimum of 15,000, more preferably 18,000, and even more preferably 20,000. The maximum is preferably 40,000, and more preferably 35,000. If the number-average molecular weight is less than 15,000, the strength and elongation of the sheet is inadequate, due to inadequate resin cohesion, which makes it brittle and unusable. On the other hand, because the melt viscosity rises if the number-average molecular weight is 40,000 or above, the optimal temperature for calendering also rises, which results in poor roll release properties in the sheet.

[0025] The acid value of the polyester resin of the present invention used for calendering is preferably 60 eq/10⁶ g or lower, more preferably 50 eq/10⁶ g or lower, and even more preferably 40 eq/10⁶ g or lower. If the acid value is more than 60 eq/10⁶ g, hydrolysis is further accelerated when the resin is heated during calendering, and the mechanical strength of the finished sheet is reduced. The roll release properties of the sheet also decline as decomposition of the resin progresses.

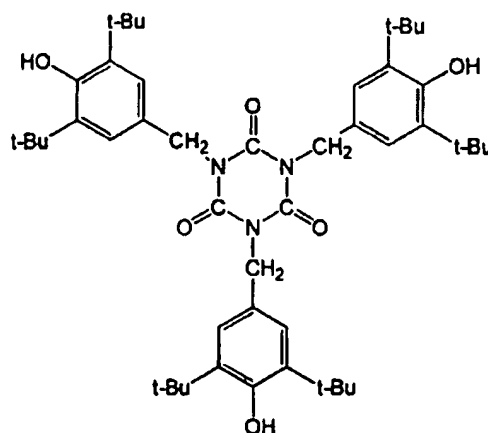
[0026] The melt viscosity of the polyester resin used in the present invention is preferably 6,000 to 60,000 dPa·sec when the temperature is 220°C and the shear speed is 100 sec⁻¹, more preferably 7,000 dPa·sec or above, and even more preferably 8,000 dPa·sec or above; the maximum is more preferably 50,000 dPa·sec or below, and even more preferably 40,000 dPa·sec

or below. If the melt viscosity is less than 6,000 dPa·sec, the roll release properties of the sheet decline due to increased adhesion of the resin. On the other hand, a melt viscosity of over 60,000 dPa·sec is too high, and the resultant decrease in productivity makes it impractical.

[0027] When a sheet is made by means of calendering from the polyester resin of the present invention, an antioxidant must be admixed with the polyester resin in order to improve the takeoff properties of the molten sheet. Among several antioxidants, the one having effects in takeoff properties consists of the tris(3,5-di-*t*-butyl-4-hydroxybenzyl)isocyanurate indicated by Formula I. By means of the hindered phenol-type antioxidant having the structure indicated by Formula I, extremely significant effects can be obtained relating to roll takeoff properties.

[0028] Formula I

[Chemical Formula 3]



[0029] The minimum admixed quantity of the hindered phenol-type antioxidant in Formula I should be 0.01 weight parts, preferably 0.02 weight parts, and more preferably 0.05 weight parts. The maximum admixed quantity should be 2 weight parts, preferably 1.8 weight parts, and more preferably 1.5 weight parts. If the admixed quantity is less than 0.01 weight parts, there may be no significant effects in the takeoff properties of the sheet from the rolls. On the other hand, if the admixed quantity is more than 2 weight parts, the compatibility of the non-crystalline polyester declines, and adverse effects on the outer appearance of the sheet can occur whereby the transparency of the sheet is compromised, and bleed-out of the antioxidant occurs when the sheet is used for an extended period.

[0030] It is more preferable to admix a thioether-type antioxidant in the polyester resin composition of the present invention for the purpose of improving the takeoff properties of the

2-tert-butyl-4,6-dimethylphenol, 2,4,6-tri-tert-butylphenol, 2-tert-butyl-4-methoxyphenol, 3-methyl-4-isopropylphenol, 2,6-di-tert-butyl-4-hydroxymethylphenol, 2,2-bis(4-hydroxyphenyl)propane, bis(5-tert-butyl-4-hydroxy-2-methylphenyl)sulfide, 2,5-di-tert-amylhydroquinone, 2,5-di-tert-butylhydroquinone, 1,1-bis(3-tert-butyl-4-hydroxy-5-methylphenyl)butane, bis(3-tert-butyl-2-hydroxy-5-methylphenyl)methane, 2,6-bis(2-hydroxy-3-tert-butyl-5-methylbenzyl)-4-methylphenol, bis(3-tert-butyl-4-hydroxy-5-methylbenzyl)sulfide, bis(3-tert-butyl-5-ethyl-2-hydroxyphenyl)methane, bis(3,5-di-tert-butyl-4-hydroxydiphenyl)methane, bis(3-tert-butyl-2-hydroxy-5-methylphenyl)sulfide, 1,1-bis(4-hydroxyphenyl)cyclohexane, ethylene bis[3,3-bis(3-tert-butyl-4-hydroxyphenyl)butyrate], bis[2-(2-hydroxy-3-tert-butyl-5-methylbenzyl)-4-methyl-6-tert-butylphenyl]terephthalate, 1,1-bis(2-hydroxy-3,5-dimethylphenyl)-2-methylpropane, 4-methoxyphenol, cyclohexylphenol, p-phenylphenol, catechol, hydroquinone, 4-tert-butylpyrocatechol, ethyl gallate, propyl gallate, octyl gallate, lauryl gallate, cetyl gallate, β -naphthol, 2,4,5-trihydroxybutyrophenone, 1,3,5-trimethyl-2,4,6-tris(3,5-di-tert-butyl-4-hydroxybenzyl)benzene, 1,6-bis[2-(3,5-di-tert-butyl-4-hydroxyphenyl)propionyloxy]hexane, tetrakis[3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionyloxymethyl]methane, bis(3-cyclohexyl-2-hydroxy-5-methylphenyl)methane, bis[3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionyloxyethyl]sulfide, n-octadecyl²-3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate, bis[3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionylamino]hexane, 2,6-bis(3-tert-butyl-2-hydroxy-5-methylphenyl)-4-methylphenol, bis[S-(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)]thioterephthalate, 1,1,3-tris(3-tert-butyl-4-hydroxy-6-methylphenyl)butane, and the like. Also, one, two, or more types of these compounds may be used together.

[0036] The admixed quantity of the phenol-type antioxidant is preferably a maximum of 1.0 weight parts or less, and particularly preferably 0.8 weight parts or less; and a preferred minimum is 0.01 weight parts or more, and particularly preferably 0.02 weight parts or more. If the admixed quantity is less than 0.01 weight parts, thermal degradation during fabrication is difficult to minimize in an efficient manner, and if the admixed quantity is more than 1.0 weight parts, the effects that minimize thermal degradation are excessive, which is uneconomical.

[0037] Specific examples of organic phosphite ester compounds used in the present invention include triphenyl phosphite, tris(methylphenyl) phosphite, triisooctyl phosphite, tridecyl phosphite, tris(2-ethylhexyl) phosphite, tris(nonylphenyl) phosphite, tris(octylphenyl) phosphite,

² Translator's note: The original reads, "otadecyl," which appears to be a typographical error.

tris[decylpoly(oxyethylene)] phosphite, tris(cyclohexylphenyl) phosphite, tricyclohexyl phosphite, tri(decyl)thio phosphite, tsiisodecyl thiophosphite, phenyl-bis(2-ethylhexyl) phosphite, phenyl-diisodecyl phosphite, tetradecyl poly(oxyethylene)-bis(ethylphenyl) phosphite, phenyl-dicyclohexyl phosphite, phenyl-diisooctyl phosphite, phenyl-di(tridecyl) phosphite, diphenyl-cyclohexyl phosphite, diphenyl-isooctyl phosphite, diphenyl-2-ethylhexyl phosphite, diphenyl-isodecyl phosphite, diphenyl-cyclohexyl phenyl phosphite, diphenyl-(tridecyl)thio phosphite, nonylphenyl-ditridecyl phosphite, phenyl-p-tert-butylphenyl-dodecyl phosphite, diisopropyl phosphite, bis[o[c]tadecyl poly(oxyethylene)] phosphite, octylpoly(oxypropylene)-tridecyl poly(oxypropylene) phosphite, monoisopropyl phosphite, diisodecyl phosphite, diisooctyl phosphite, monoisooctyl phosphite, didodecyl phosphite, monododecyl phosphite, dicyclohexyl phosphite, monocyclohexyl phosphite, monododecyl poly(oxyethylene) phosphite, bis(cyclohexylphenyl) phosphite, monocyclohexyl-phenyl phosphite, bis(p-tert-butylphenyl) phosphite, tetratridecyl-4,4'-isopropylidene diphenyl diphosphite, tetratridecyl 4,4'-butylidene bis(2-tert-butyl-5-methylphenyl) diphosphite, tetraisooctyl-4,4'-thiobis(2-tert-butyl-5-methylphenyl) diphosphite, tetrakis(nonylphenyl)-poly(propyleneoxy)isopropyl diphosphite, tetratridecyl-propyleneoxypropyl diphosphite, tetratridecyl-4,4'-isopropylidene-dicyclohexyl diphosphite, pentakis(nonylphenyl)-bis[poly(propyleneoxy)isopropyl] triphosphite, heptakis(nonylphenyl)-tetrakis[poly(propyleneoxy)isopropyl] pentaphosphite, heptakis(nonylphenyl) tetrakis(4,4'-isopropylidenediphenyl) pentaphosphite, decakis(nonylphenyl)-heptakis(propyleneoxyisopropyl) octaphosphite, decaphenyl-heptakis(propyleneoxyisopropyl) octaphosphite, bis(butoxycarboethyl)-2,2-dimethylene-trimethylene dithiophosphite, bis(isooctoxycarbomethyl)-2,2-dimethylene trimethylene dithiophosphite, tetradecyl-ethylene dithiophosphite, tetradodecyl-hexamethylene dithiophosphite, tetradodecyl-2,2'-oxydiethylene dithiophosphite, pentadodecyl-di(hexamethylene) trithiophosphite, diphenyl phosphite, 4,4'-isopropylidene-dicyclohexyl phosphite, 4,4'-isopropylidenediphenyl-alkyl(C12 to C15) phosphite, 2-tert-butyl-4-[1-(3-tert-butyl-4-hydroxyphenyl)isopropyl]phenyl di(p-nonylphenyl) phosphite, ditridecyl-4,4'-butylidene bis(3-methyl-6-tert-butylphenyl) phosphite, dioctadecyl-2,2-dimethylene trimethylene diphosphite, tris(cyclohexylphenyl) phosphite, hexatridecyl-4,4',4''-1,1,3-butanetriyl-tris(2-tert-butyl-5-methylphenyl) triphosphite, tridodecyl thiophosphite, decaphenyl-heptakis(propyleneoxyisopropyl) octaphosphite³, dibutyl-pentakis

³ Translator's note: The original reads, "octabosphite," which appears to be a typographical error.

(2,2-dimethylene trimethylene) diphosphite, dioctyl-pentakis(2,2-dimethylene trimethylene) diphosphite, didecyl-2,2-dimethylene trimethylene diphosphite, and metal salts thereof with lithium, sodium, potassium, magnesium, calcium, barium, zinc, and aluminum. Also, one, two, or more types of these compounds may be used together.

[0038] The admixed quantity of the organic phosphite ester compound is preferably a maximum of 3.0 weight parts or less, and particularly preferably 2.0 weight parts or less; and a preferred minimum is 0.01 weight parts or more, and particularly preferably 0.02 weight parts or more. If the admixed quantity is less than 0.01 weight parts, thermal degradation during fabrication is difficult to minimize in an efficient manner, and if the admixed quantity is more than 3.0 weight parts, the effects that minimize thermal degradation are excessive, which is uneconomical.

[0039] When a sheet is made by means of calendering from the polyester resin of the present invention, a lubricant must be admixed with the polyester resin in order to improve the roll release properties of the molten sheet.

[0040] The admixed quantity of the lubricant used in the present invention is 0.01 to 5 weight parts. A preferred minimum is 0.05 weight parts, a more preferred minimum is 0.1 weight parts, and an even more preferred minimum is 0.2 weight parts. Also, a preferred maximum is 4.5 weight parts, a more preferred maximum is 4 weight parts, and an even more preferred maximum is 3.5 weight parts. If the quantity of the lubricant is less than 0.01 weight parts, enhancement to the roll release properties is difficult to obtain, and if the quantity is more than 5 weight parts, the transparency, coloration, and printability of the processed sheet show a tendency to decline.

[0041] The lubricants used in the present invention include polyolefinic waxes, metal salts of organic phosphate esters, organic phosphate esters, ester compounds of adipic acid or azelaic acid with higher aliphatic alcohols, ethylene bis-stearamides, methylene bis-stearamides, ester compounds of glycerin with higher aliphatic acids, ester compounds of pentaerythritol with higher aliphatic acids, higher aliphatic alcohols, higher aliphatic acids, paraffins derived from petroleum or coal, waxes, natural or synthetic polyester waxes, metallic soaps made from higher aliphatic acids, and the like. These may be used in combinations of one, two, or more types thereof. Using a polyolefinic wax and/or a metal salt of an organic phosphate ester is particularly preferred, because the roll release properties of the sheet and the transparency of the finished sheet can be easily obtained at the same time.

[0042] Polyolefinic waxes used as lubricants in the present invention include polyethylene wax, polypropylene wax, and derivatives of these, and these derivatives include copolymers with other monomers, such as acrylic acid, vinyl acetate, styrene, and maleic acid, and partially oxidized decomposition products.

[0043] Metal salts of organic phosphate esters used as lubricants in the present invention include, for example, metal salts of the organic phosphate ester indicated by the general formula (III) below and/or metal salts of the organic phosphate ester indicated by the general formula (IV) below.

[0044] Formula (III): $[\{RO(C_fH_{2f}O)_n\}_{3-a-o}PO(O)_a(OH)_e]_d\{M(OH)_b\}_c$

(In the formula, R is a hydrocarbon group with a carbon number of 4 to 30; M is an alkali metal, alkaline earth metal, Zn, or Al; a is 1 or 2; e is 0 or 1 (0 or 1 when a is 1, and 0 when a is 2); b is 0 to 2; c is 1 or 2; d is 1 to 3; f is 2 or 3; n is 0 to 60; and a, b, c, and d have the following relation with the valence (hereinafter referred to as m) of the metal (M). When m = 1, then b = 0, d = 1, and a = c; when m = 2, then b = 0, c = 1, and a × d = 2, or b = 1, d = 1, and a = c; when m = 3, then b = 0, d = 3, a = c, b = 1, c = 1 and a × d = 2, or b = 2, d = 1, and a = c; furthermore, when m ≥ 2, mutually different phosphate ion groups may be bonded with the metal (M), and in this case, d = 2 or 3 is the total number of mutually different phosphate ion groups. Also, when d is 2 or 3, each of the structures enclosed in brackets may be mutually the same or different.)

[0045] Formula (IV): $[\{R^1O(C_fH_{2f}O)_n\}_{3-a-e}PO(O)_a(OH)_e]_d\{M(OCOR^2)_s(OH)_x\}_t$

(In the formula, R¹ is a hydrocarbon group with a carbon number of 4 to 30; R² is an alkyl group with a carbon number of 1 to 25; M is an alkali metal, alkaline earth metal, Zn, or Al; a is 1 or 2; e is 0 or 1 (0 or 1 when a is 1, and 0 when a is 2); d is 1 or 2; s is 1 or 2; x is 0 or 1; t is 1 or 2; f is 2 or 3; and n is 0 to 60. Also, s + x = 1 or 2; and a, d, s, and t have the following relation with the valence (hereinafter referred to as m) of the metal (M). When m = 2, then s = 1, d = 1, and a = t; when m = 3, then s = 1, t = 1, and a × d = 2, or s = 2, d = 1, and a = t; and furthermore, when m = 3, mutually different phosphate ion groups may be bonded with the metal (M), and in this case, d = 2 is the total number of various phosphate ion groups. Also, when d is 2, each of the structures enclosed in brackets may be mutually the same or different.)

[0046] An alkyl group, phenyl group, arylalkyl group, alkenyl⁴ group, or alkylphenyl group is preferred for the hydrocarbon group with a carbon number of 4 to 30 indicated by R in the abovementioned general formula (III) and the hydrocarbon group with a carbon number of 4 to 30 indicated by R¹ in the abovementioned general formula (IV). Also, Li, Na, K, and the like, for example, are preferred for the alkali metal indicated by M in the general formulae (III) and (IV), and Mg, Ca, Ba, and the like, for example, are preferred as alkaline earth metals.

[0047] The metal salt of the organic phosphate ester indicated by the general formula (III) and the metal salt of the organic phosphate ester indicated by the general formula (IV) may be manufactured by means of the usual method, and no particular limit is placed on the manufacturing method.

[0048] Preferred examples of metal salts of the organic phosphate ester indicated by the general formula (III) include chemical compounds (1) through (13) in Table 1 below, chemical compounds (14) through (16) in Table 2 below, and the like, and preferred examples of metal salts of the organic phosphate ester indicated by the general formula (IV) include chemical compounds (15) through (26) in Table 2 below. These may be used singly or together in combinations of two or more types thereof.

[0049] Also, as shown by decimal fractions included in the number of repetitions of oxyethylene or oxytrimethylene units in the polyethyl alcohol component, these compounds (compounds (1) through (26)) are pure compounds or mixtures of a plurality of metal salts of phosphate esters having different numbers of repeating oxyethylene units or oxytrimethylene units in the polyether alcohol component (the number of repetitions (n) in (C₁H₂₁O)_n in the formula).

⁴ Translator's note: The original has "argenyl," which appears to be a typographical error.

[0050]

[Table 1]

Compound No.	Structural Formula	Compound No.	Structural Formula
(1)	$\text{C}_{16}\text{H}_{33}(\text{OCH}_2\text{CH}_2)_{28}\text{O}-\text{P}(=\text{O})(\text{OH})\text{ONa}$	(9)	$\text{C}_9\text{H}_{19}(\text{OCH}_2\text{CH}_2)_{103}\text{O}-\text{P}(=\text{O})(\text{OH})\text{ONa}$
(2)	$\text{C}_{18}\text{H}_{37}(\text{OCH}_2\text{CH}_2)_{28}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OLi}$	(10)	$\text{C}_{20}\text{H}_{41}(\text{OCH}_2\text{CH}_2)_{49}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OLi}$
(3)	$\text{C}_{18}\text{H}_{37}(\text{OCH}_2\text{CH}_2)_{163}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OLi}$		$\text{C}_{20}\text{H}_{41}(\text{OCH}_2\text{CH}_2)_{43}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OLi}$
(4)	$\text{C}_{13}\text{H}_{27}(\text{OCH}_2\text{CH}_2)_{64}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OMgOH}$	(11)	$\text{C}_{10}\text{H}_{21}(\text{OCH}_2\text{CH}_2)_{103}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OMg}$
(5)	$\text{C}_{14}\text{H}_{29}(\text{OCH}_2\text{CH}_2)_{43}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OLi}$		$\text{C}_{18}\text{H}_{37}(\text{OCH}_2\text{CH}_2)_{43}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OLi}$
(6)	$\text{C}_{12}\text{H}_{25}(\text{OCH}_2\text{CH}_2)_{73}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OK}$	(12)	$\text{C}_{18}\text{H}_{35}(\text{OCH}_2\text{CH}_2)_{343}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OLi}$
(7)	$\text{C}_{13}\text{H}_{27}(\text{OCH}_2\text{CH}_2)_{59}\text{O}-\text{P}(=\text{O})(\text{OH})\text{Zn}$	(13)	$\text{C}_{13}\text{H}_{27}(\text{OCH}_2\text{CH}_2)_{69}\text{O}-\text{P}(=\text{O})(\text{OH})\text{Zn}$
(8)	$\text{C}_{13}\text{H}_{27}(\text{OCH}_2\text{CH}_2)_{69}\text{O}-\text{P}(=\text{O})(\text{OH})\text{Zn}$		

[0051]

[Table 2]

Compound No.	Structural Formula	Compound No.	Structural Formula
(14)	$\text{C}_{18}\text{H}_{37}(\text{OCH}_2\text{CH}_2\text{CH}_2)_{5.4}\text{O}-\text{P}(=\text{O})(\text{OH})\text{ONa}$	(21)	$\text{C}_{12}\text{H}_{25}(\text{OCH}_2\text{CH}_2)_{6.3}\text{O}-\text{P}(=\text{O})(\text{O})_2-\text{Al}-\text{C}_7\text{H}_{15}\text{COO}$
(15)	$\left[\left(\text{C}_9\text{H}_{19} \right)_2 \text{C}_6\text{H}_4 (\text{OCH}_2\text{CH}_2)_{5.2} \text{O} \right]_2 \text{O}-\text{P}(=\text{O})(\text{OH})_2-\text{Ca}$	(22)	$\left[\text{C}_{13}\text{H}_{27}(\text{OCH}_2\text{CH}_2)_{7.0} \right]_2 \text{O}-\text{P}(=\text{O})(\text{O})_2-\text{CH}_3\text{COO}-\text{AlOH}$
(16)	$\text{C}_{24}\text{H}_{49}\text{O}-\text{P}(=\text{O})(\text{OH})_2-\text{Zn}$	(23)	$\left[\text{C}_{18}\text{H}_{33}(\text{OCH}_2\text{CH}_2)_{6.3} \right]_2 \text{O}-\text{P}(=\text{O})(\text{O})_2-\text{C}_7\text{H}_{13}\text{COO}-\text{Zn}$
(17)	$\text{C}_{14}\text{H}_{29}(\text{OCH}_2\text{CH}_2)_{7.8} \text{O}-\text{P}(=\text{O})(\text{OH})_2-\text{Ba}-\text{O}-\text{C}_{17}\text{H}_{35}\text{COO}$	(24)	$\text{C}_6\text{H}_5(\text{OCH}_2\text{CH}_2)_{18.5} \text{O}-\text{P}(=\text{O})(\text{OH})_2-\text{C}_8\text{H}_{17}\text{COO}$
(18)	$\text{C}_9\text{H}_{19}(\text{OCH}_2\text{CH}_2)_{5.0} \text{O}-\text{P}(=\text{O})(\text{OH})_2-\text{Mg}-\text{O}-\text{C}_{11}\text{H}_{23}\text{COO}$	(25)	$\text{C}_{18}\text{H}_{37}(\text{OCH}_2\text{CH}_2\text{CH}_2)_{11.2} \text{O}-\text{P}(=\text{O})(\text{OH})_2-\text{Ba}-\text{O}-\text{C}_{17}\text{H}_{35}\text{COO}$
(19)	$\text{C}_8\text{H}_{17}(\text{OCH}_2\text{CH}_2)_{18.5} \text{O}-\text{P}(=\text{O})(\text{OH})_2-\text{Ca}-\text{O}-\text{C}_7\text{H}_{15}\text{COO}$	(26)	$\left(\text{C}_{15}\text{H}_{31}\text{O} \right)_2 \text{O}-\text{P}(=\text{O})(\text{O})_2-\text{C}_{11}\text{H}_{23}\text{COO}-\text{Zn}$
(20)	$\left[\text{C}_{13}\text{H}_{27}(\text{OCH}_2\text{CH}_2)_{7.0} \right]_2 \text{O}-\text{P}(=\text{O})(\text{O})_2-\text{C}_{11}\text{H}_{23}\text{COO}-\text{Zn}$		

In the polyester resin composition of the present invention, other components may be arbitrarily added according to the application. These components include, for example, fillers, UV absorbers, light stabilizers, pigments, antistatic agents, antimicrobial agents, epoxy⁵ compounds, cross-linking agents, and the like.

[0052] The aforementioned components of the polyester resin composition of the present invention used for calendering are mixed using a Henschel mixer or other known mixing and stirring device, and the mixture is kneaded with the help of a Banbury mixer, mixing roll, warming roll, or the like, and calendered.

[0053] The roll temperature during calendering of the polyester resin composition of the present invention is not particularly limited, but a temperature of about 160 to 200°C is preferred, and about 170 to 190°C is particularly preferred.

[0054] The polyester resin composition of the present invention possesses excellent roll release properties, whereby the adhesion time (time from the beginning of kneading until the composition substantially adheres to the heating roll) to the roll heated to 180°C during calendering is less than 15 minutes.

[0055] Also, the polyester composition of the present invention has a haze of 15% or lower when calendered to a thickness of 0.7 mm, which is a calendered quality (high transparency of a fabricated sheet) that was unachievable using a conventional polyester resin composition.

[0056] The thickness of a sheet obtained by means of calendering the polyester resin composition⁶ of the present invention varies according to the application, but is generally 10 to 1,000 µm, preferably 30 to 800 µm. Also, the "sheet" referred to herein is conceptually the same as a thin product often generally referred to as a "film."

[0057] The sheet of the present invention is suitable for use in shrink labeling used for food products, cosmetics, and beverages, as well as in aroma-retaining heat sealed film, oil-resistant multilayer sheeting, sheeting used in building materials, sheeting used for food packaging, blister pack sheeting, sheeting used in writing materials, and the like, and is suitable for building material sheeting, food container sheeting, blister pack sheeting, and sheeting for writing materials, in which particularly high transparency is required. Also, aside from having high

⁵ Translator's note: The word "epoxy" reads "cboxy" in the original, which appears to be a typographical error. This is corrected in the translation for the sake of readability.

⁶ Translator's note: The word "composition" contains an erroneous character in the original, which appears to be a typographical error. This word is corrected in the translation for the sake of readability.

transparency, the use of non-crystalline polyester confers the product with the advantages of being easily plasticized and comparatively easy to work, which makes it useful in printed decorative sheeting.

[0058]

[Working Examples] The following working examples will be given in order to describe the present invention in further detail, but the present invention is in no way limited by the working examples. Measured values given in the synthesis examples were found by means of the measurement methods below.

Resin composition: The product was dissolved in heavy chloroform, and assayed by means of H-NMR.

Glass transition temperature: Measured using a differential scanning calorimeter by placing a 10-mg measurement sample on an aluminum pan, pressing the lid closed, and measuring at a temperature increase rate of 20°C/min.

Number-average molecular weight: Found as a polystyrene equivalent value by gel permeation chromatography using hexafluoroisopropanol as a solvent.

Acid value: 1 g of the resin was dissolved in 30 mL of chloroform, and the acid value was found by titration with a 0.1 N potassium hydroxide ethanol solution. Phenolphthalein was used as an indicator.

[0059] <Synthesis example of non-crystalline polyester (A)>

960 weight parts of dimethyl terephthalate, 580 weight parts of ethylene glycol, 170 weight parts of neopentyl glycol, and 0.34 weight part of tetrabutyl titanate were placed in a reactor equipped with a stirrer, thermometer, and an effluent cooler, and transesterification was performed for two hours at 170 to 220°C. As the temperature of the reaction system was raised from 220°C to 270°C after completion of transesterification, the pressure inside the system was gradually reduced to 500 Pa over 60 minutes. Polycondensation was then performed for 50 minutes at 130 Pa or below, and non-crystalline polyester (A) was obtained.

[0060] According to NMR analysis of non-crystalline polyester (A), the dicarboxylic acid component was composed of 100 mol% of terephthalic acid, and the diol component was composed of 75 mol% of ethylene glycol and 25 mol% of neopentyl glycol. Also, the glass transition temperature thereof was 78°C, the number-average molecular weight was 26,000, and the acid value was 27 eq/10⁶ g. Non-crystalline polyesters (B) through (E) were manufactured in

the same manner as non-crystalline polyester (A). The composition and results of measurement thereof are shown in Table 3. (Numerical values are mol% in resin.)

[0061]

[Table 3]

		Non-crystalline polyester synthesis examples				
		A	B	C	D	E
Acid	Terephthalic acid	100	90	88	85	100
	Isophthalic acid		10		15	
	Adipic acid			12		
Glycol	Ethylene glycol	75	85	78	72	71
	Diethylene glycol		15			
	Neopentyl glycol	25				
	Cyclohexanedimethanol					29
	2-methyl-1,3-propanediol			22		
	1,3-propanediol				28	
Properties	Number-average molecular weight	28000	32000	25000	33000	29000
	Glass transition temperature (°C)	78	62	55	58	78
	Melting point (°C)	—	—	—	—	—
	Acid value (equivalent weight/10 ⁶ g)	30	25	16	38	25

[0062] <Working Examples and Comparative Examples>

The polyesters shown in Table 3 were mixed in a beaker with the components shown in Tables 4 and 5, and the mixture was kneaded on two 6-inch chilled rolls set to 180°C. The product was mixed while occasionally peeling off the resin adhering to the chilled rolls, and after five minutes of kneading, the roll interval was set to 0.3 mm (sheet thickness set to 0.3 mm), the molten sheet was taken off the roll to a distance of 30 cm, and the sheet takeoff properties were evaluated by visual observation of its sagging when taken off. The release properties of the sheet from the roll at that time were also evaluated. Also, the evaluation criteria were as shown below.

[0063] Sheet release properties:

○: Good release properties from the roll

×: Strong adhesion to the roll, releasing difficult, normal sheet cannot be obtained

Sheet takeoff properties:

◎: No sagging at all

○: Slight sagging, but to no effect in practical use

×: Molten sheet sags under its own weight, normal sheet cannot be obtained

[0064] The transparency of the sheet was also evaluated as follows. The same kneading as described above was performed using two test rolls of the same product used in the abovementioned evaluation of sheet takeoff properties and sheet release properties (roll release properties), the roll interval was set to 0.5 mm after five minutes, and a sheet having a thickness of 0.5 mm was obtained. In order to increase the smoothness of the surface of the sheet and even out the thickness thereof, the sheet thus obtained was cut to a size of 13 cm×13 cm, and two layers thereof were stacked and sandwiched between ferrotype plates (stainless hard chrome). The assembly was then pressed and cooled under conditions of 180°C, 90 seconds, and 12 N/cm², and an evaluation sample was obtained having a thickness of 0.7 mm.

Haze measurement: The abovementioned evaluation sample was cut to a size of 3 cm×3 cm to make a test sample, which was measured using an ND-Σ80 manufactured by Nippon Electronic Industries, Ltd. The smaller the haze value the better the transparency, and a haze of 15% or below was considered acceptable (○), and a haze of over 15% was considered unacceptable (×).

[0065] Results thereof are shown in Tables 4 and 5. In the section for evaluation of takeoff properties and transparency, "-" indicates that evaluation was not performed because the release properties of the sheet were poor and a normal sheet could not be obtained.

[0066] Also, the lubricants and plasticizers indicated in Tables 4 and 5 refer to the compounds below.

a: tris(3,5-di-t-butyl-4-hydroxybenzyl)isocyanurate

b: pentaerythritol tetrakis(3-laurylthiopropionate)

c: tetrakis[methylene-3-(3,5-di-t-butyl-4-hydroxyphenyl)propionate]methane

d: distearyl-3,3'-thiodipropionate

e: zinc tridecyl poly(oxyethylene) phosphate

f: styrene-modified polyethylene wax

Also, the numerical values for the quantities of polyesters, antioxidants, and lubricants in the tables represent weight parts.

[0067]

[Table 4]

		Working Examples								
		1	2	3	4	5	6	7	8	9
Non-crystalline polyester	A	100	100	100					100	
	B				100					
	C					100				
	D							100		
	E						100			100
Stabilizer	a	0.3	0.3	0.5	0.5	0.2	0.5	0.3	0.5	0.3
	b		0.1	0.2			0.1			
	c									0.1
	d								0.1	
Lubricant	e	0.5	0.5	1.0	1.0	0.8	0.5	0.8	0.8	0.5
	f	0.1	0.1	0.2	0.2	0.2	0.1	0.2	0.2	0.1
Evaluation	Sheet release properties	○	○	○	○	○	○	○	○	○
	Sheet takeoff properties	○	◎	◎	○	○	◎	○	○	○
	Transparency	○	○	○	○	○	○	○	○	○

[0068]

[Table 5]

		Comparative Examples						
		1	2	3	4	5	6	7
Non-crystalline polyester	A	100	100	100	100			
	B							
	C							
	D						100	
	E					100		100
Stabilizer	a			0.3				
	b						0.2	0.2
	c				0.3	0.2		0.5
	d					0.1		
Lubricant	e		0.5		0.5	0.5	0.8	1.0
	f		0.1		0.1	0.1	0.2	0.2
Evaluation	Sheet release properties	×	○	×	○	○	○	○
	Sheet takeoff properties	—	×	—	×	×	×	×
	Transparency	—	○	—	○	×	○	×

[0069]

[Effect of the Invention] As described above, the polyester resin composition of the present invention consists of a composition in which non-crystalline polyester, an antioxidant, and a lubricant are combined, and therefore allows the molten sheet to be readily taken off from the rolls during calendering of the polyester resin composition, which was difficult in the past, and enables sheet calendering in which the finished sheet has good transparency and excellent quality. Also, because the sheet is provided with the good physical properties of the polyester, has a high degree of transparency, and contains no plasticizers that have agglomerated impurities therein, it is highly safe, and because it also uses non-crystalline polyester, subsequent processing of the sheet thus fabricated is relatively easy.

Continued from first page:

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